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Comparison of phenomenological models with a microscopic theory for semiconductor optical nonlinearities

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Abstract. A microscopic theory of the third order coherent semiconductor optical response is used for the investigation of four-wave mixing signals. The theory is based on the dynamics-controlled truncation formalism and evaluated for the case of resonant excitation of heavy-hole excitons. This approach allows for a direct comparison with other theories (microscopic or phenomenological), and yields a detailed understanding of important many-body effects such as excitation-induced dephasing.

Introduction

The nonlinear optical response of semiconductors quantum wells has long been an important aspect of both theoretical and experimental semiconductor physics [1, 3]. The understanding of many-body effects such as excitonic effects and charge carrier-correlations is important for a correct interpretation of nonlinear optical response of quantum wells. Important examples of recent experiments include the observation of biexcitonic beatings in four-wave mixing (FWM) experiments [2] and strong polarization dependence of the FWM signal in semiconductor microcavities [4].

The theoretical description of optically excited semiconductors is usually based on various modifications and extensions of the so-called semiconductor Bloch equations, which are equations of motion for the wavevector-dependent interband polarization function and the charge carrier (i.e. electron and hole) distribution functions. The simplest approximation is the Hartree–Fock (HF) approximation, in which all correlation functions are factorized in terms of one-particle functions.

However, there exist several theoretical approaches for dealing with many-body processes beyond the HF approximation. One of them is the dynamics-controlled truncation (DCT) formalism, which is a rigorous approach in, for example, the $\chi^{(3)}$ nonlinear regime if one restricts the analysis to the coherent limit with zero electron and hole density initial conditions [5]. These equations can be written in terms of the interband polarization function and the two-exciton correlation function. If consistent with the excitation conditions, the equations can be restricted to contain only optically excited 1s-excitons. In that case it is advantageous to expand all functions in terms of exciton functions and keep only the smallest set of excitons, that is consistent with the excitation conditions [7].

While the numerical complications only allow for an inclusion of a few eigenstates (1s in our case), this is still well justified approximation for a resonant excitation, especially in semiconductor with large exciton binding energies are used or spectrally narrow pulses (e.g. picosecond pulses). From this approach one can extract and calculate the nonlinear optical response function and extract different quantities related to many-body interaction effects, e.g. phase-space filling, Hartree–Fock terms, coherent biexciton contributions and exciton continuum scattering contributions. These quantities can be directly compared with

the phenomenological parameters entering other models such as weakly interacting Boson (WIB) model (see e.g. [6]) or various few-level models. The comparison with the WIB model is done elsewhere [8], while the comparison with few-level models is discussed in this papers.

1. Theory and discussion

In this section we discuss both the microscopic theory based on the DCT formalism as well as the 3rd-order nonlinear optical response based on a commonly used phenomenological five-level model.

The five-level system corresponds to two two-level systems representing the heavy holes in the angular momentum states $|\pm 3/2>$, the electrons in the spin states $|\pm 1/2>$, and a phenomenologically included biexciton state.

Using an atomic density matrix approach one can write the general equation of motion for the density operator in the form

$$i\hbar\frac{d}{dt}\rho_{mn} = (\hbar\omega_{mn} - i\gamma'\sum_{i>g}\rho_{ii})\rho_{mn} + \sum_{i}(H'_{ni}\rho_{im} - \rho_{ni}H'_{im}), \tag{1}$$

where the interaction hamiltonian $H'_{mn} = -\vec{\mu_{mn}}\cdot(\vec{E} + L\vec{P})$, $\vec{\mu}_{mn}$ is the dipole matrix element, $\vec{E}$ is the optical field, and $\vec{P}$ is the spin-resolved polarization function. The many-body effects are accounted for by the inclusion of the biexciton level, the excitation-induced dephasing (EID) associated with the parameter γ', and the local field factor L. Iterating this equation to the third order one can derive the expression for the total polarization using $\vec{p} = -eTr(\vec{r}\rho) = \sum_{nm}\vec{\mu}_{nm}\rho_{mn}$. For the spin $+1$ polarization component one obtains

$$\begin{aligned}
P_+^{(3)} \propto\ & \frac{-i}{\hbar}\int_{t_0}^{t} dt' e^{-i\omega_x(t-t')}\{-i\gamma'\rho_+(t')[|\rho_+(t')|^2 + |\rho_-(t')|^2] \\
& + E_+(t')2|\rho_+(t')|^2 + \tilde{L}\rho_+(t')2|\rho_+(t')|^2 \\
& - \frac{i|\beta|^2}{\hbar}\int_{t_0}^{t'} dt'' e^{-i(2\omega_x-\delta_{bx})(t'-t'')}\{E_-^*(t')[E_+(t'')\rho_-(t'') + E_-(t'')\rho_+(t'')] \\
& + \tilde{L}\rho_-^*(t')[E_+(t'')\rho_-(t'') + E_-(t'')\rho_+(t'')] \\
& + \tilde{L}2E_-^*(t')\rho_+(t'')\rho_-(t'') + \tilde{L}^2 2\rho_-^*(t')\rho_+(t'')\rho_-(t'')]\} \\
& \frac{-i|\beta|^2}{\hbar}\int_{t_0}^{t} dt' e^{-i(\omega_x-\delta_{bx})(t-t')}\{ \\
& -\rho_-^*(t')[E_+(t')\rho_-(t') + E_-(t')\rho_+(t')] - \tilde{L}2\rho_-^*(t')\rho_+(t')\rho_-(t') \\
& + \frac{i}{\hbar}\int_{t_0}^{t'} dt'' e^{-i(2\omega_x-\delta_{bx})(t'-t'')}\{E_-^*(t')[E_+(t'')\rho_-(t'') + E_-(t'')\rho_+(t'')] \\
& + \tilde{L}\rho_-^*(t')[E_+(t'')\rho_-(t'') + E_-(t'')\rho_+(t'')] \\
& + 2\tilde{L}E_-^*(t')\rho_+(t'')\rho_-(t'') + 2\tilde{L}^2\rho_-^*(t')\rho_+(t'')\rho_-(t'')\}\}.
\end{aligned} \tag{2}$$

A similar expression can be derived for the spin -1 polarization component. We use the notation $\rho_\pm$ to denote the first order density matrix (linear optical response) $E_\pm = \vec{\mu}_\pm\vec{E}$, $\tilde{L} = L|\mu|^2/V$, where V is the volume or, as in our case of a quasi-two dimensional system, the total area of the system.

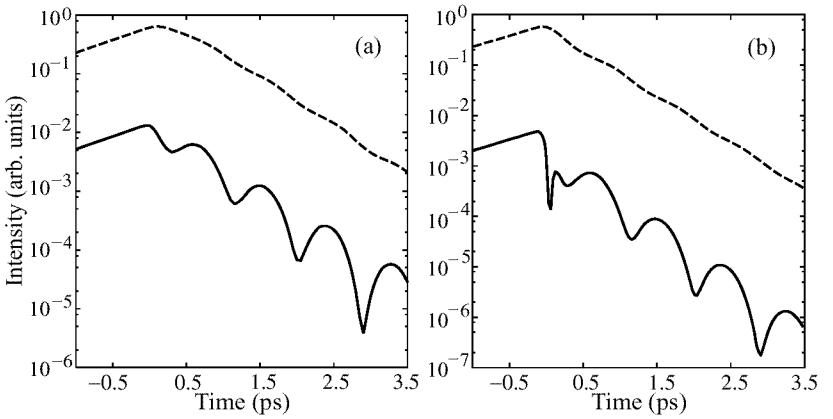

Fig. 1. Four-wave mixing signal vs pump-probe delay time in co-polarized (dashed lines) and cross-polarized (solid line) configuration computed with the microscopic (a) and phenomenoligical (b) theory.

We now turn to the microscopic formulation. The equation for the third-order polarization that one can be obtain from the DCT equations in terms of exciton wave functions can be written in the following form:

$$
\begin{aligned}
p_+^{(3)}(t) = \frac{-i}{\hbar}\int_{t_0}^{t} dt' \mathrm{e}^{-i\omega_x(t-t')}\{2C_{1s}E_+(t')|p_+(t')|^2 + V^{st}p_+(t')|p_+(t')|^2 \\
-\frac{i}{\hbar}\int_{t_0}^{t'} dt''\{\mathrm{e}^{-i(2\omega_x-\delta_{bx})(t'-t'')} f_0 p_-^*(t')p_+(t'')p_-(t'') \\
+2f^+(t'-t'')p_+^*(t')p_+(t'')p_+(t'') \\
+[f^+(t'-t'') + f^-(t'-t'')]p_-^*(t')p_+(t'')p_-(t'')\}\}. \qquad (3)
\end{aligned}
$$

Here, p is the first order coherence function. The two terms on the first line of this equation represent the phase space filling (PSF) and Hartree–Fock (HF) contributions. This is followed by the bound biexciton term on the 2nd line, (restricted to the singlet channel) and the rest of the equation contains the continuum scattering terms, with contributions in both triplet and singlet channels. Here "singlet" (f_0 and f^-) and "triplet" (f^+) refer to the electron spin states. The definitions of the parameters C_{1s} and V^{st} (see, e.g., [10]), as well as f_0, and $f^{\pm}(t'-t'')$ are lengthy and not given here, they include multiple integrals with exciton eigenfunctions, the Coulomb potential. The biexciton contribution and continuum contribution in the singlet channel contain also the biexciton wave function (which can be found in Ref. [11]).

Equations (3) and (2) are a suitable basis for a detailed comparison of parameter entering the phenomenological model with the interaction terms derived from the microscopic theory. Whereas a strict one-to-one correspondence does not exist, one can indeed identify approximate correspondences and, thus, validate to a certain extent the phenomenological model. As far as numerical agreement is concerned, we show in Fig. 1 results for time-integrated four-wave mixing signal in a ZnSe semiconductor quantum well.

In conclusion, we present a theoretical analysis of ultrafast optical nonlinearties in semiconductors. We believe that our detailed comparison of phenomenological model

with a microscopic theory is important in order to validate conclusions about physical processes obtained from the widely used phenomenological models.

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